

# Q-Factor Matching for Femtosecond Near-Field Enhancement in Hybrid Metal-Dielectric Metasurfaces

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Near-field enhancement in nanocavities governs the efficiency of nonlinear optical processes and ultrafast light-matter interactions. However, maximizing the cavity quality factor does not necessarily maximize the response under femtosecond excitation. Here, we experimentally demonstrate that hybrid metal-dielectric metasurfaces provide a practical platform for optimizing this trade-off. By coupling a low-Q localized surface plasmon resonance of Au nanodisks to a high-Q dielectric mode of a TiO<sub>2</sub> metasurface, we continuously tune the Q factors of the hybrid modes over a broad range while preserving the plasmonic hotspot geometry. Using four-photon photoemission electron microscopy under 100-fs excitation, we map the nonlinear near-field response and correlate it with spectrally extracted Q factors and ultrafast dynamics measured by time-resolved photoemission electron microscopy. The response varies nonmonotonically with Q and reaches a maximum at  $Q \approx 20$ , where the photoemission yield is enhanced approximately 15-fold relative to the uncoupled metasurface. These results identify pulse-cavity Q-factor matching between the cavity and the driving pulse as a key design principle for pulsed-laser nanophotonics.

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Nanocavities that support strong near-field mode confinement are central to the enhancement of nonlinear optical processes and ultrafast light-matter interactions [1–10]. Metal nanostructures are especially attractive because localized surface plasmon resonances (LSPRs) can generate extremely small mode volumes and intense hotspots at material interfaces, which are favorable for hot-carrier generation, photochemistry, and interface photocurrent conversion [1–4]. Their practical limitation, however, is the intrinsically low Q factor imposed by strong radiative and nonradiative damping [11–13]. In contrast, dielectric metasurfaces can support much higher-Q resonances, including Mie resonances and

quasibound states in the continuum [5–9], but typically with weaker subwavelength localization unless special gap geometries are introduced [14–16].

Hybrid metal-dielectric metasurfaces provide a route to combine these complementary advantages [17–22]. In such systems, the plasmonic component preserves intense near-field localization, whereas the dielectric component suppresses radiative decay and extends the lifetime of the hybrid modes. Importantly, this architecture makes it possible to tune the hybrid-mode Q factor over a broad range while keeping the plasmonic building block nearly unchanged. This offers a clean platform for boosting the near field and addressing a fundamental question for resonant nanocavities driven far from the continuous-wave limit: what cavity Q maximizes the nonlinear near-field response under ultrashort-pulse excitation?

In this Letter, we use hybrid Au-TiO<sub>2</sub> metasurfaces to address this question directly. By varying the  $x$ -axis lattice constant  $P_x$ , we tune the coupling between an Au LSPR and the transverse magnetic (TM) mode of a TiO<sub>2</sub> metasurface from the weakly coupled regime to pronounced

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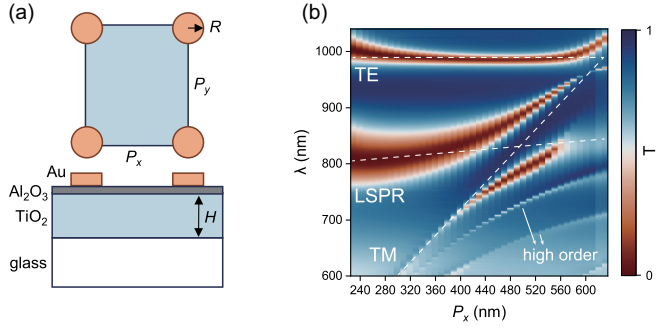


FIG. 1. Design of hybrid metal-dielectric metasurface. (a) Schematic of the structure. Designed structural parameters: lattice periods  $P_x$  and  $P_y$ ,  $\text{TiO}_2$  film thickness  $H = 170$  nm, Au disk radius  $R = 75$  nm, Au disk height 40 nm, and  $\text{Al}_2\text{O}_3$  spacer thickness 10 nm. The substrate is glass-coated with a thin ITO layer ( $\sim 10$  nm). (b) Simulated transmittance spectra under  $x$ -polarized normal-incidence illumination as  $P_x$  is varied. A broad plasmonic mode and two narrow dielectric modes are observed and labeled LSPR, TE, and TM, respectively. The LSPR-TM hybridization enables wide tuning of the hybrid-mode Q factors, whereas the TE mode remains spectrally separated. Two higher-order dielectric modes are also indicated.

hybridization. We quantify the resulting response using wavelength-resolved four-photon photoemission electron microscopy (PEEM) under 100-fs excitation, extract Q

factors from normal-incidence transmittance spectra, and determine the ultrafast dephasing dynamics with time-resolved PEEM (TR-PEEM). The measured PEEM signal, which serves as an experimental metric for the pulsed nonlinear near-field response [23–36], first increases and then decreases with increasing Q. The optimum occurs at  $Q \approx 20$ , well below the equivalent spectral Q factor of the driving pulse. Here, Q-factor matching does not mean equality between the cavity Q and the spectral Q of the laser pulse. Rather, it denotes the optimum balance among field buildup, coupling efficiency, pulse bandwidth, temporal overlap, and the nonlinear order of the detected process. Our Letter presents a strategy for boosting near-field enhancement under pulsed lasers and highlights the importance of Q-factor selection in practical applications.

The sample design is shown in Fig. 1(a), with periodic Au nanodisks on a  $\text{TiO}_2$  film. The Au nanodisks support LSPRs and simultaneously act as couplers to excite the dielectric metasurface modes. The 10-nm  $\text{Al}_2\text{O}_3$  spacer serves as a matching layer to lower the local refractive index surrounding the Au nanodisks. This allows the Au nanodisks to be designed considerably larger than those directly on  $\text{TiO}_2$  for improved structural homogeneity. Figure 1(b) shows representative simulated transmittance spectra obtained by sweeping  $P_x$  while keeping  $P_y$  and the Au-disk geometry fixed under  $x$ -polarized normal-incidence

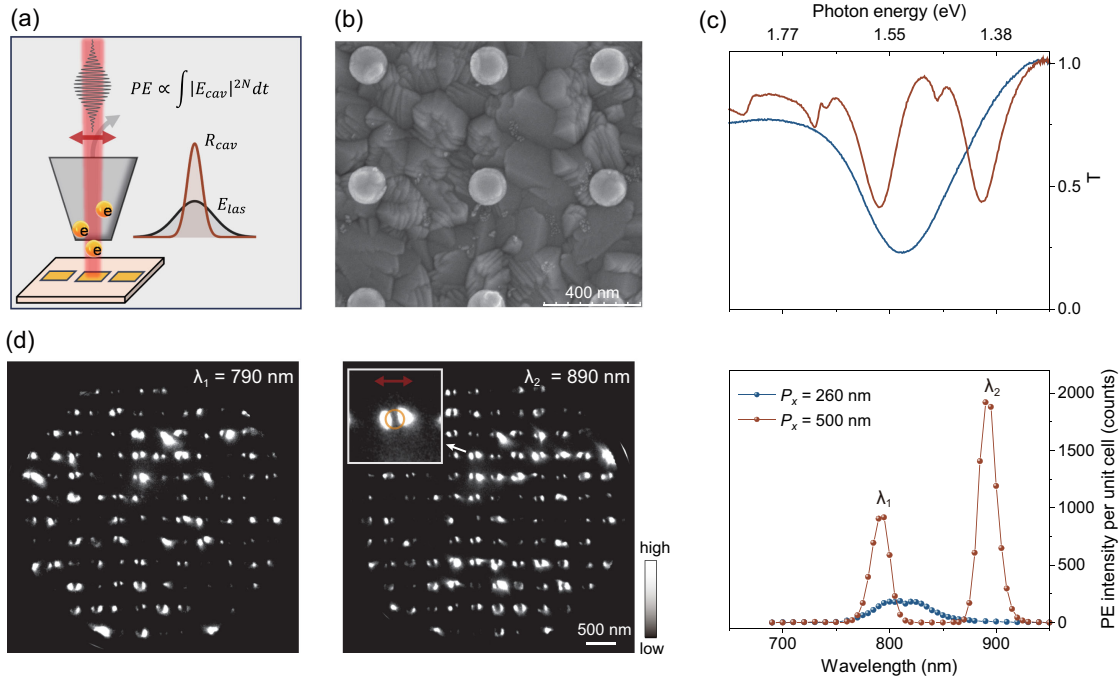


FIG. 2. Near-field characterization of the hybrid modes by PEEM. (a) Schematic of the PEEM experiments using a normally incident 100-fs tunable laser. The photoemission intensity is directly correlated to the near-field intensity. (b) Representative scanning electron microscopy image of a metasurface with  $P_x = 500$  nm, fixed  $P_y = 500$  nm, and  $R = 77.5$  nm. (c) Transmittance spectra (upper) and integrated photoemission intensity spectra (lower) of two metasurfaces corresponding to the uncoupled case ( $P_x = 260$  nm) and a strongly hybridized case ( $P_x = 500$  nm). (d) Corresponding PEEM images of the  $P_x = 500$  nm metasurface at the two hybrid-mode peaks. The inset enlarges the hot spot distribution around a single Au disk. The PEEM signal was integrated over the entire  $60 \times 60 \mu\text{m}$  metasurface array and normalized by the number of unit cells.

illumination. A broad LSPR and two narrow dielectric modes are observed, denoted transverse electric (TE) and TM. The TE mode is deliberately kept spectrally separated, whereas the TM mode hybridizes strongly with the LSPR. As a result, the hybrid-mode linewidths and Q factors can be controlled efficiently through  $P_x$  over a wide range. As discussed in Supplemental Material [37], the dielectric TE and TM resonances are associated with quasibound states in the continuum modes of the  $\text{TiO}_2$  metasurface (corresponding to the case of central disk  $R_1 = 0$ ) [38,39].

To probe the pulsed near-field enhancement experimentally, we performed wavelength-resolved PEEM measurements using a 100-fs tunable laser incident normally on the sample, as illustrated in Fig. 2(a). The incident polarization was aligned along the  $x$  axis. In the near infrared, PEEM relies on multiphoton photoemission to overcome the work function [27,32] and has become a powerful tool for mapping plasmonic near fields in space and time. The photoemission intensity follows  $P_E \propto I^N$ , where  $P_E$  is the photoemission intensity,  $I$  is the local light intensity ( $I \propto |E_{\text{loc}}|^2$ ), and  $N$  is the nonlinear order. Here,  $N$  is around 4.0, representing a four-photon process. Figure 2(b) shows a representative scanning electron microscopy image of a metasurface array at  $P_x = 500$  nm, in which the modes are at the strong coupling regime [4]. The corresponding transmittance spectrum in Fig. 2(c) (upper panel) exhibits two hybrid resonances at 790 and 890 nm. The PEEM

images acquired at these wavelengths, shown in Fig. 2(d), reveal hot spots concentrated at the opposite edges of the Au disks along the  $x$  axis, consistent with the expected plasmonic localization. By sweeping the laser wavelength at fixed incident power, we obtain the integrated PEEM signal per unit cell [Fig. 2(c), lower panel], which we use as an experimental metric of the pulsed nonlinear near-field enhancement, denoted by  $PE \propto \int |E_{\text{cav}}|^{2N} dt$ . Relative to the uncoupled metasurface ( $P_x = 260$  nm), the coupled sample exhibits both higher Q factors and substantially stronger photoemission intensity. Quantitatively, the Q factors of the two hybrid peaks are 20.5 and 25.5, whereas the uncoupled LSPR has  $Q = 7.6$ ; the corresponding peak PEEM yields are enhanced by factors of about 5.6 and 10.7, respectively.

To characterize the ultrafast dynamics of the hybrid modes, we further performed TR-PEEM measurements. A 7.0-fs source spanning 670–950 nm was used in an interferometric pump-probe configuration based on a Mach-Zehnder interferometer. Because the TE mode lies near 1000 nm, outside this spectral window, it does not contribute to the measured TR-PEEM response. We estimated the time-dependent local electric field  $E(t)$  by fitting the interferometric traces with a coupled-mode model [40,41], as described in Supplemental Material [37]. Figure 3 shows that the hybridized modes exhibit substantially longer dephasing times than the uncoupled LSPR, in agreement with the spectral narrowing seen in the transmission measurements. Specifically, the dephasing times extracted from  $E(t)$  are about 8 fs for the uncoupled metasurface and 22 and 30 fs for the two hybrid modes of the  $P_x = 500$  nm sample.

The full  $P_x$ -dependent evolution is summarized in Fig. 4, including transmittance spectra, photoemission intensity spectra, and Q factors. As shown in Figs. 4(a) and 4(b), the photoemission spectra track the same resonance wavelengths as the transmittance spectra, but their intensity values vary largely as the coupling condition changes. As  $P_x$  increases, the system evolves from the uncoupled regime to pronounced hybridization and then back toward weaker interaction. For both the lower and upper branches in photon energy domain, the photoemission intensity first increases and then decreases with  $P_x$ . As shown in Fig. 4(c), the lower branch reaches its maximum near  $P_x = 440$  nm, whereas the upper branch peaks near  $P_x = 520$  nm. By contrast, the Q factor evolves monotonically along each branch: it increases for the lower branch and decreases for the upper branch. When the PEEM response is plotted directly against Q [Fig. 4(d)], both branches collapse onto the same central message: the pulsed nonlinear near-field response is maximized at an intermediate Q of approximately 20 rather than at the highest accessible Q. For the lower branch, the maximum enhancement is approximately 15-fold relative to the uncoupled metasurfaces with  $P_x = 240$  nm during the full

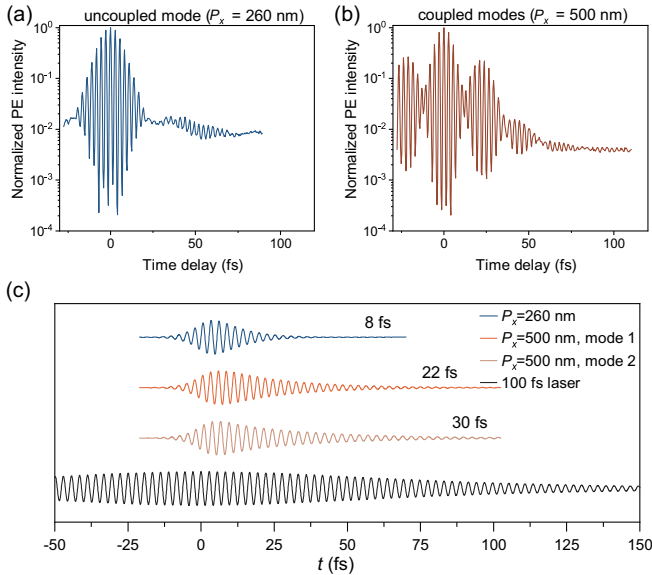


FIG. 3. Ultrafast dynamics of the hybrid modes measured by TR-PEEM. (a),(b) Interferometric TR-PEEM traces for the uncoupled metasurface and the hybridized metasurface, respectively. The modulation in (b) results from the beating between the two excited modes. (c) Estimated time-domain electric-field oscillations  $E(t)$ . For comparison, the envelope of the 100-fs pulse is also shown. Modes 1 and 2 correspond to the resonances at 790 and 890 nm, respectively.

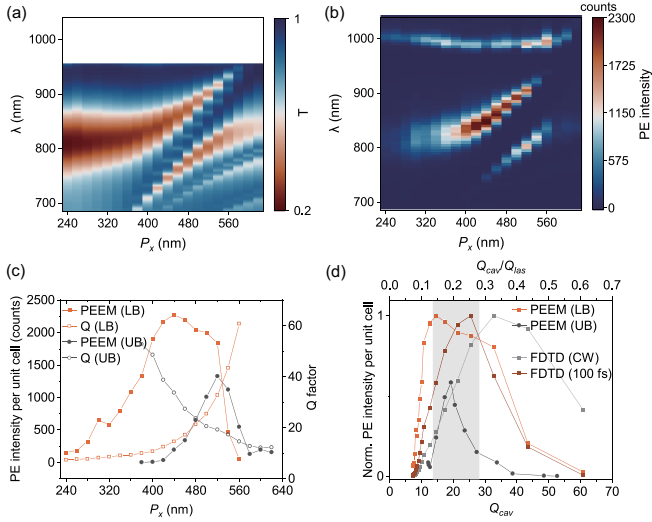


FIG. 4. Summary of the  $P_x$ -dependent measurements, including transmittance spectra, photoemission intensity spectra, and Q factors. (a) Transmittance spectra; no data is shown above 950 nm because of the detector range. (b) Integrated PEEM spectra acquired by sweeping the laser wavelength at fixed incident power. (c) PEEM peak intensity and Q factor as functions of  $P_x$ . (d) Normalized PEEM peak intensity plotted against Q factor. The upper horizontal axis shows  $Q_{cav}/Q_{las}$ , where  $Q_{las} = 100$  is the equivalent spectral Q factor of the 100-fs excitation pulse. For the lower branch, the FDTD simulation results that represent CW and 100-fs pulse excitations are shown. The optimal Q factor shifts from approximately  $Q \sim 35$  in the CW-FDTD simulation to approximately  $Q \sim 25$  in the 100-fs FDTD simulation, which is closer to the experimental optimum around  $Q \sim 20$ . LB, lower branch; UB, upper branch.

$P_x$ -dependent sweeping. The peaks are broader for the lower branch compared to the upper branch. This difference is likely because the lower branch has much cleaner modes, while the upper branch is affected by nearby higher-order dielectric modes. Additional discussions on  $P_x$ -sweeping and strong-coupling analysis are provided in Supplemental Material [37].

Our experiments demonstrate the boost of near-field enhancement by hybrid metal-dielectric metasurfaces under pulsed laser. The physics of this behavior can be understood in two steps. We first consider the continuous-wave (CW) limit and then include the finite duration of the femtosecond driving pulse. In the CW picture, a single-mode resonator stores energy efficiently when the radiative and absorptive loss channels are balanced. This critical-coupling principle provides the correct starting point for understanding why a bare LSPR, which is typically over-coupled and radiatively lossy, does not deliver the maximum possible field enhancement despite its extremely small mode volume.

For a single resonant mode driven under CW excitation, the steady-state field enhancement can be written as Eq. (1) (see Sec. VI of Supplemental Material [37]) [42,43],

$$K^2 = \frac{|E|^2}{|E_0|^2} \propto \frac{2\gamma_{rad}}{(\gamma_{rad} + \gamma_{abs})^2 V_{eff}} \propto \frac{4Q_{tot}^2}{Q_{rad} V_{eff}}. \quad (1)$$

Here,  $\gamma_{rad}$  and  $\gamma_{abs}$  denote the radiative and absorption damping rates, respectively.  $Q = \omega_0/2\gamma$  and the total Q factor fulfills  $Q_{tot}^{-1} = Q_{rad}^{-1} + Q_{abs}^{-1}$ ;  $V_{eff}$  is the mode volume. At critical coupling ( $\gamma_{rad} = \gamma_{abs}$ ), Eq. (1) gives the maximum field enhancement,

$$K_{max}^2 \propto \frac{Q_{abs}}{V_{eff}}. \quad (2)$$

Bare plasmonic resonators generally operate in the over-coupled regime with  $\gamma_{rad} \gg \gamma_{abs}$ , so their actual field enhancement  $K_{LSPR}^2 \propto (4Q_{rad}/V_{eff})$  remains well below the critical-coupling limit. For a coupled-mode system, the final hybrid modes are coherent superposition of the two components. The single-mode Eq. (2) is no longer directly applicable, but its underlying principle remains instructive. Hybridization with the dielectric mode suppresses radiative leakage and increases the effective Q factor while inheriting the strongly localized plasmonic hot spot. Because the mode volume varies only weakly over the relevant  $P_x$  range according to the simulations in Supplemental Material [37], the dominant control parameter in the present system is the hybrid-mode Q factor.

Consider the lower branch, for example. This picture explains the initial rise of the PEEM signal as the lower branch evolves away from the uncoupled plasmonic resonance. For  $240 \text{ nm} \leq P_x \leq 440 \text{ nm}$ , the mode remains predominantly plasmonic while Q increases, leading to stronger near-field response. As  $P_x$  increases further, the hybrid mode acquires a larger dielectric fraction and shifts substantially in wavelength; the PEEM response then decreases even though Q continues to rise. Frequency-domain finite-difference time-domain (FDTD) simulations in the CW limit reproduce the overall trend, but they place the optimum at a higher Q ( $\sim 35$ ) and predict a larger enhancement ratio ( $\sim 10^2$ ) than observed experimentally. The discrepancy points directly to the role of the finite pulse duration and to experimental factors such as sample inhomogeneity and hot spot to hot spot morphological variations. Although sample inhomogeneity can partly explain the discrepancy in enhancement ratio and peak broadening, it is unlikely to fully account for the systematic shift of the optimal response toward lower Q.

The femtosecond driving pulse provides the missing ingredient. The local field  $E_{loc}(t)$  is the convolution of the 100-fs laser field with the response function of the cavity mode. As Q increases, the spectral linewidth narrows and the temporal response lengthens, reducing the overlap between the cavity response and the finite-bandwidth laser pulse in both frequency and time. Consequently, the excitation efficiency no longer increases monotonically with Q. Time-domain simulations that explicitly include a

100-fs source reproduce this shift and move the optimum closer to the experimental value. An analytical Q-matching model presented in Supplemental Material [37] captures the same trend and shows that, for nonlinear observables, the optimal cavity Q can be substantially smaller than the equivalent spectral Q of the driving pulse.

Taken together, the PEEM measurements, TR-PEEM dynamics, and pulse-aware modeling show that the relevant design principle for femtosecond nanophotonics is not simply to maximize Q, but to optimize the cavity response for the finite duration and bandwidth of the excitation pulse. In the present four-photon experiment, this matching condition places the optimum near  $Q \approx 20$ , far below the highest-Q modes accessible in the structure. The exact optimum will depend on the pulse duration, spectral profile, and nonlinear order, but the underlying principle is general.

The present platform also suggests routes toward larger responses. Because the hybrid modes retain plasmonic hot spots, further enhancement should be possible through optimized metal geometry—for example, by introducing sharper features or nanogaps [32] while maintaining the moderate-Q hybridization condition identified here. More broadly, the measured correlation between nonlinear PEEM response and cavity Q provides an experimentally grounded benchmark for designing metasurfaces for harmonic generation, hot-carrier processes, and other ultrafast light-matter interactions driven by short pulses [44]. For example, these hybrid structures can potentially be applied to photoionization and strong-field optics, where carrier-envelope phase and dispersion effects can be further explored.

In conclusion, we have experimentally demonstrated Q-factor matching for femtosecond near-field enhancement in hybrid metal-dielectric metasurfaces. By tuning the coupling between an Au LSPR and a dielectric  $\text{TiO}_2$  mode, we realize a controllable set of hybrid resonances and directly correlate their PEEM yields, Q factors, and ultrafast dephasing dynamics. The optimum response occurs at  $Q \approx 20$  and yields an enhancement of approximately 15-fold relative to the uncoupled metasurface. These results identify Q-factor matching as a practical design rule for pulsed-laser nanophotonics and highlight hybrid metal-dielectric metasurfaces as a versatile platform for enhancing ultrafast nonlinear phenomena.

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**Data availability**—There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

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